

N72-30469

NASA CONTRACTOR
REPORT

NASA CR-61391

CASE FILE
COPY

BANDING DUE TO TEMPERATURE OSCILLATIONS
IN THE UNIDIRECTIONAL SOLIDIFICATION
OF EUTECTIC ALLOYS


By A. Kidron
Hayes International Corporation
Huntsville, Alabama

August 1, 1972

Prepared for

NASA-GEORGE C. MARSHALL SPACE FLIGHT CENTER
Marshall Space Flight Center, Alabama 35812

TECHNICAL REPORT STANDARD TITLE PAGE

1. REPORT NO. NASA CR-61391		2. GOVERNMENT ACCESSION NO.		3. RECIPIENT'S CATALOG NO.	
4. TITLE AND SUBTITLE BANDING DUE TO TEMPERATURE OSCILLATIONS IN THE UNIDIRECTIONAL SOLIDIFICATION OF EUTECTIC ALLOYS				5. REPORT DATE August 1, 1972	
				6. PERFORMING ORGANIZATION CODE	
7. AUTHOR(S) A. Kidron				8. PERFORMING ORGANIZATION REPORT # WD-551	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Hayes International Corporation Huntsville, Alabama				10. WORK UNIT NO.	
				11. CONTRACT OR GRANT NO.	
				13. TYPE OF REPORT & PERIOD COVERED CONTRACTOR REPORT	
12. SPONSORING AGENCY NAME AND ADDRESS National Aeronautics and Space Administration Washington, D. C. 20546				14. SPONSORING AGENCY CODE	
15. SUPPLEMENTARY NOTES					
16. ABSTRACT <p style="text-align: center;">Banding observed in unidirectional solidification of eutectic alloys is shown to be due to melting back of the freezing interface because of oscillations in the temperature of the furnace. General theoretical criteria as to the amplitude and frequency of the permissible temperature oscillations are given to ensure that banding will not occur.</p>					
17. KEY WORDS			18. DISTRIBUTION STATEMENT Unclassified - unlimited		
					
19. SECURITY CLASSIF. (of this report) U		20. SECURITY CLASSIF. (of this page) U		21. NO. OF PAGES 12	
				22. PRICE \$3.00	

INTRODUCTION

Bands have been observed during unidirectional solidification of eutectic Al Cu alloys. These bands are almost planar and perpendicular to the growth direction which is along the long axis of the solidified ingot (Figure 1). Such bands had actually been seen elsewhere ⁽¹⁾ and were interpreted as due to convection currents in the liquid metal because of changes in the density of the solidifying metal.

A closer look at the temperature - time charts in the present case showed that in the cases where these bands were seen the temperature was not kept constant but showed some oscillations. It was then important to see if these oscillations were actually the reason for banding which could then be eliminated or if convection currents were the reason for the banding so that a different approach would have to be used to eliminate the banding.

To this end experimental observations were made which showed that actually the fluctuations in the temperature were the reason for banding. A theoretical analysis was then carried out to calculate the amplitude and frequency of the permissible temperature oscillations which would not produce any banding.

Experimental Observations

The bands have been observed in a few different samples. A typical one is the G1-2-T2-2 sample, micrographs of which are given in Figure 1. The wavelength of the bands is seen to vary along the sample. It is about 100 microns in the cold side of the specimen and rises up to about 250 microns at the other end. A closer look into the structure of the bands with a magnification of 660X shows (Figure 1) that at the band most of the lamella stopped growing suddenly while some of them grew into a globular form. A structure similar to this one was observed by Chadwick.⁽²⁾ In the latter study single crystal of Cu Al_2 - Al eutectic alloys were obtained in which the lamella had grown parallel to the long axis of the ingot. The single crystals were partially remelted to about half their length and then allowed to resolidify. The structure of the meltback position looks very similar to the structure of the bands observed here in the sample G1-2-T2-2. We have then a process of melting back for a short duration and then regrowing of the lamella again until next meltback occurs and so on.

The power on the heating element had to be reduced at a given rate during solidification in order to achieve the required decrease of the temperature with time. To this end the power was put on and off at given intervals. The interval of time when the power was on

decreased with time while the total on-off interval was constant.

In the solidification of the G1-2-T2-2 sample the total on-off period was 19 seconds. This resulted in oscillations in the temperature of the heater with an amplitude of about $\pm 2^{\circ}\text{C}$. During former runs when no banding was observed the on-off period of the power was about 1 second. In such cases the temperature of the heater showed oscillations with an amplitude of about $\pm 0.5^{\circ}\text{C}$.

To check if the bands are related to the temperature oscillations another run was made using the sample D5-1-T3-1. This time the temperature controller was not used and instead, the temperature was reduced by lowering the voltage on the heating element at a given rate. Also, twice during the experiment, the temperature was given a small spike. Micrographs taken from this sample after solidification are given in Figure 2. One can see that no banding is visible along the ingot except at two places. The distance between the two bands is 18.6 mm.

A look at the temperature-time chart of this run shows that the two temperature spikes occur at $T=645^{\circ}\text{C}$ and at $T=597^{\circ}\text{C}$, the time interval between the spikes being of 26 minutes. This yields a cooling rate of $1.84^{\circ}/\text{min}$. Assuming that the two bands seen in the sample are due to these two temperature spikes one finds that the temperature gradient in this interval is about $25.8^{\circ}/\text{cm}$ and the growth rate is 11.8 microns/sec. or 4.2 cm/hr.

Experimentally the run with the sample D5-1-T3-1 was similar to the run of the sample G1-2-T2-2. Consequently we assume that in the latter sample the growth rate was also of 11.8 microns/sec. As mentioned above in this sample the on-off period of the heater was 19 seconds. The periodic disturbance of the temperature should then produce bands with a wavelength of $11.8 \times 19 = 224$ microns. This is similar to the wavelength of the bands observed actually near the end of the specimen (about 250 microns as mentioned above). We can then conclude that the temperature disturbances due to the temperature controller are the cause of the banding observed in the specimen G1-2-T2-2.

Theoretical Calculations

An accurate analysis of the effect of the temperature oscillations on the solidification process is complicated because of the many factors involved. Also the full amplitude of a temperature fluctuation will not be felt at a growing S/l interface, and the fluctuations will be partially absorbed and damped out by the thermal boundary layer. In pure materials such fluctuations will have only a small effect, but in alloys the solute distribution in front of the S/l interface will be greatly affected.

Several analyses of this effect have been carried out in recent years. ^(3,5) The most elaborate one is due to Hurle et al. ⁽⁴⁾ and we will use their theory to analyze our case of Al-Cu Al₂ eutectic alloy solidification.

In this theory one writes down the equations for thermal and mass flow when a planar solid-liquid phase boundary moves with constant velocity v along a given axis (z). The assumptions made are:

- (1) The flow is only in one direction, (z).
- (2) There is no mass-diffusion in the solid.
- (3) There is no kinetic undercooling of the interface.
- (4) The temperature varies linearly in the region of interest.

After the steady-state solution is obtained, the fluctuation in the temperature is introduced as a first order perturbation in the temperature and composition, so that the temperature is now

$$T = T^{(0)} + T^{(1)} \quad \text{and the composition is}$$

$$C = C^{(0)} + C^{(1)}$$

where $T^{(1)}$ and $C^{(1)}$ are the perturbations, assumed to be small. The velocity of the interface will then be

$$v = v^{(0)} + v^{(1)} = v^{(0)} + d\phi/dt$$

where ϕ and $v^{(1)}$ are measured relative to the frame of coordinates moving with the unperturbed velocity $v^{(0)}$. It must be emphasized here that while the distance $\phi(t)$ is small as a perturbation, $v^{(1)}$ is not necessarily so.

These perturbations yield new boundary conditions which are then used to solve the equations of heat and mass flow under the new assumption that the fluctuation in the temperature of the liquid is periodic with time, i.e.

$$T_l^{(1)} = \theta e^{i\omega t}$$

If melting-back occurs then the change in the concentration in the solid $C_s^{(1)}$ will be dependent on the previous freezing and will have a complicated dependence on time. So in order to obtain some solution one assumes that melting - back does not take place. Under this assumption one obtains different equations which make it possible to calculate the values of

$$\frac{1}{\theta} \left| \frac{v^{(1)}}{v^{(0)}} \right| \text{ and } \frac{1}{\theta} \left| \frac{C_s^{(1)}}{C_1^{(0)}} \right| \text{ and also of the root mean square}$$

concentration gradient versus $\frac{W}{v^{(0)}}$ where W is the frequency of the oscillations.

For the case of the Al-Cu Al_2 eutectic alloy with oscillations of a period $T = 1$ Sec. $v^{(0)} = 1.18 \cdot 10^{-3}$ cm/sec. we find that $\frac{1}{\theta} \left| \frac{v^{(1)}}{v^{(0)}} \right| \approx 1.6$, so that for $\theta \geq 0.6^\circ\text{C}$ we will have $\left| \frac{v^{(1)}}{v^{(0)}} \right| \geq 1$ i.e. melting-back will occur. For the case of $T = 19$ sec (the case where bands were seen) we obtain that melting back will occur if $\theta \geq 0.8^\circ\text{C}$. Indeed, in this case, as mentioned above, we had $\theta \approx 2^\circ\text{C}$ which produced banding due to melting-back.

We must warn here though that in the theory it was assumed that melting-back did not occur so that these numbers should not be taken too seriously. But nevertheless they should give the order of magnitude of the permissible frequencies and amplitudes of the temperature oscillations in order that melting-back should not occur.

Even when there is no remelting, bands could be produced by temperature oscillations for the following reason.⁽⁵⁾

In the unidirectional solidification of a eutectic alloy the solid forms as horizontal layers of the two resulting phases A and B, one on top of the other. These layers end as a vertical planar front which constitutes the solid/liquid interface. It is well established now ⁽⁶⁾ that the layers of A and B grow simultaneously so that in the liquid phase we have an A poor (B rich) region in front of the A phase layer and a B poor (A rich) region in front of the B phase layer. We have then a solution rich layer (A rich & B rich alternately) in front of the solid liquid interfaces.

This layer dissolves by lateral diffusion building up the solid phase, resulting in the advance of the solid liquid interface. At the same time a new layer is built constantly, making the process of solidification a continuous one.

In normal growth the solute rich boundary layer dissolves and builds up at a rate comparable to the rate of advance of the solid/liquid interface. But when temperature oscillations exist, as in our case, there will be times at which the interface is advancing more rapidly than the boundary layer is dissolving (moving). As a consequence, the solute rich layer becomes trapped in the solid and produces a band. But actually there is diffusion in the solid so that

bands which are narrow enough will diffuse out completely and be eliminated. From simple diffusion equations one finds that (7) bands with a wavelength of the order of 100 microns or smaller will be eliminated by solid state diffusion.

Given the rate of growth of 12 microns/sec. we will then expect that temperature oscillations of periods less than eight seconds will produce bands which will be eliminated by diffusion. This is true though if there is no melt-back, i.e. if the temperature oscillations have amplitudes smaller than $\pm 0.6^{\circ}\text{C}$. Otherwise, as we have seen above, we will have a structural morphology completely different from parallel lamella so that the probability that solid state diffusion will rehabilitate the parallel lamella becomes very small.

More generally, we will expect to obtain parallel lamella for rates of growth roughly between 1 cm/hr to 50 cm/hr for the normally used temperature gradients. At higher freezing rates the lamella degenerate into rods when some impurities are present in the melt^(8,9,10) and at low freezing rates we obtain some spheroidization.⁽¹¹⁾ This implies that to obtain lamellar eutectic solidification without banding we must have a control of any temperature oscillations such that their amplitude is smaller than about $\pm 0.5^{\circ}\text{C}$ and their frequency higher than about 2 cycles/sec.

REFERENCES

- 1) D. T. J. Hurle, in "Crystal Growth", Pergamon, Oxford, 1967, p. 659.
- 2) G. A. Chadwick, Prog. Materials Sci., 10, 124 (1963).
- 3) W. R. Wilcox and L. D. Fullmer, J. Appl. Phys, 36, 221 (1965).
- 4) D. T. J. Hurle, E. Jakeman & E. R. Pike, J. Crystal Growth, 3, 633 (1968).
- 5) G. S. Cole, in "Solidification", ASM, Cleveland, Ohio, 1971, p. 226.
- 6) L. M. Hogan, R. W. Kraft & F. D. Lemkey, in Adv. Materials Research, Vol. 5, 83 (1971).
- 7) H. P. Utech & M. C. Flemings, in "Crystal Growth", Pergamon, Oxford, 1967, p. 651.
- 8) J. D. Hunt & J. P. Chilton, J. Inst. Metals, 91, 338, (1962).
- 9) G. A. Chadwick, J. Inst. Metals, 91, 298 (1962).
- 10) A. S. Yue, J. Inst. Metals, 92, 248 (1963).
- 11) G. A. Chadwick, J. Inst. Metals, 91, 169 (1963); 92, 18 (1963).

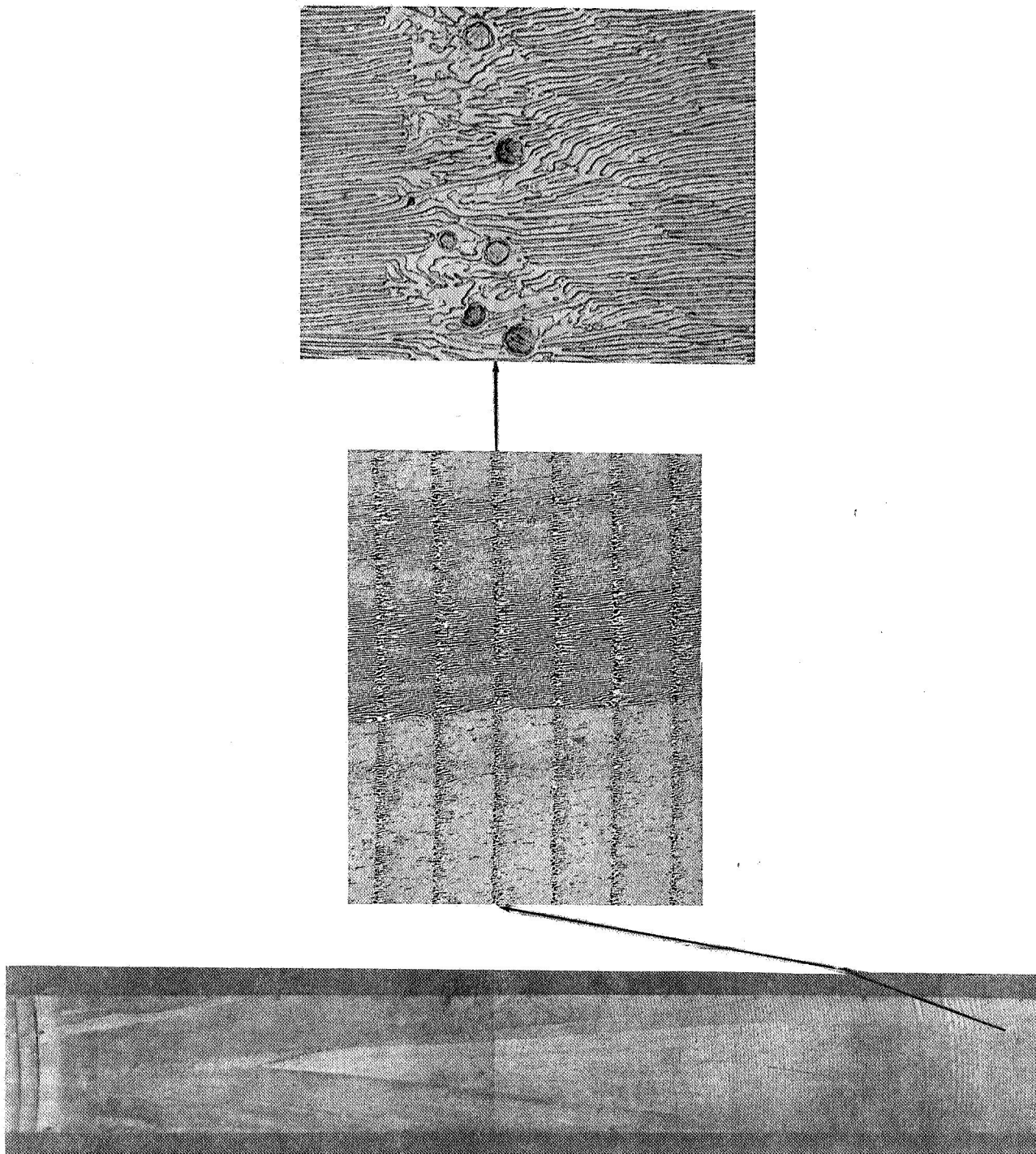
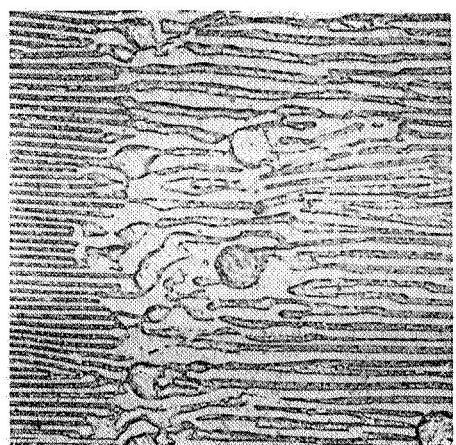
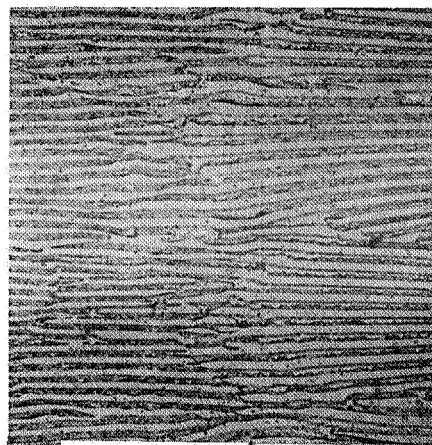


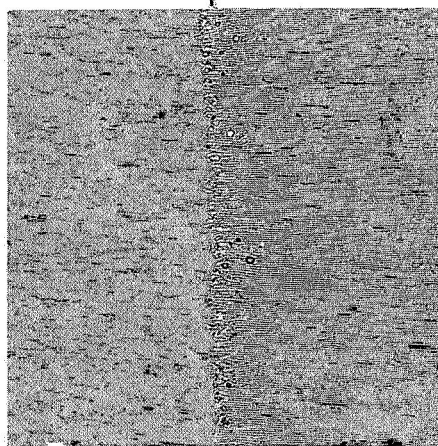
Figure 1. Banding in unidirectionally solidified Al-Cu eutectic alloy with temperature oscillations.



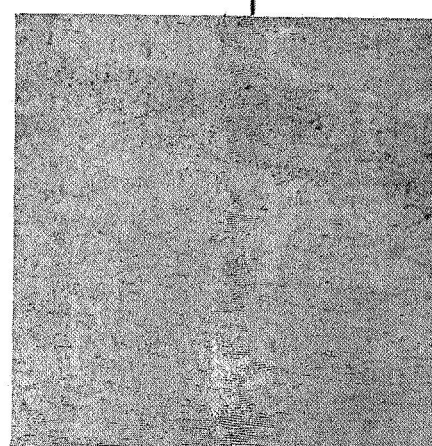
650X



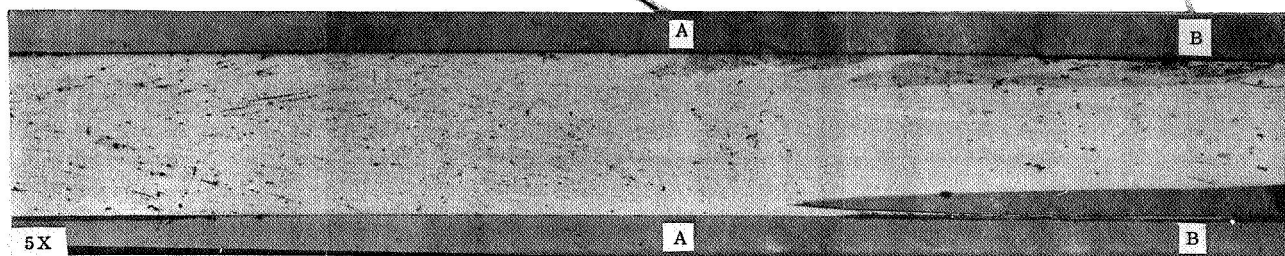
650X



80X



80X



5X

A

B

A

B

Figure 2. Unidirectionally solidified Al-Cu eutectic alloy with no temperature oscillations except for two intentional spikes at A and B